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- (S) Thermoplastic resin composition.
- A novel thermoplastic composition having improved paint adhesion strength comprising,
  - A) from about 5 to 95 weight % of at least one polyphenylene ether.
  - B) from about 95 to 5 weight % of at least one polyamide,
  - C) paint adhesion improving amount of at least one paint adhesion improver selected from the class consisting of
    - a) homopolymers or copolymers of unsaturated nitrile compounds and
  - b) homopolymers or copolymers of alkyl (meth)acrylate, excluding an ethylene alkylacrylate copolymer having about 50 weight % or higher of ethylene,
  - D) from about 0.01 to 30 parts by weight of at least one compatibilizer, based on 100 by weight of the total of the polyphenylene ether and polyamide.

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#### THERMOPLASTIC RESIN COMPOSITION

The present invention relates to novel polyphenylene ether-polyamide blends having improved paint adhesion. Specifically, polyphenylene ether polyamide blends containing at least one paint adhesion improver selected from the class consisting of

- a) homopolymers or copolymers of unsaturated nitrile compounds and,
- b) alkyl arcylate or methacrylate homopolymers or copolymers as hereinafter defined, have improved paint adhesion as compared to polyphenylene ether polyamide blends without such an improver.

Blends of polyphenylene ether and polyamide have long been known. U.S.P. 3,379,792 taught improved processability of polyphenylene ethers by incorporating therein up to 25 % by weight of polyamide.

Recently compatibility of such blends has been enhanced by incorporating therein a compatibilizing agent. U.S.P. 4,315,086 teaches the use of liquid diene polymers, epoxy compounds and compounds having in the molecule both (x) a carbon-carbon double bond or a carbon-carbon triple bond and (y) a carboxylic acid, acid anhydride, acid amide, imide, carboxylic acid ester, amino or hydroxyl group as compatibilizers. EP 46040 teaches copolymers of vinyl aromatic compounds and either an alpha, beta-unsaturated dicarboxylic acid anhydride or an imide compound thereof as a compatibilizer.

USP 4,659,763 teaches the use of quinone compounds, USP 4,600,741 teaches the use of trimellitic anhydride acid chloride and the like, USP 4,659,760 teaches the use of oxidized polyethylene wax, WO 85/05372 teaches the use of polycarboxylic acids such as citric acid, WO 87/07281 teaches the use of vinyltrimethoxy silane as compatibilizers.

It is also known to add various rubbery polymers and copolymers to further enhance the impact strength of the foregoing compatibilized polyphenylene ether/polyamide blends.

While the foregoing compatibilized and rubber modified blends exhibit good mechanical properties, their paint adhesion are not entirely suitable for certain paint systems especially with no primer coating layer applied to a substrate fabricated from those blends. It is an object of the present invention to provide polyphenylene ether polyamide composition of enhanced paint adhesion.

## SUMMARY OF THE INVENTION

According to the present invention, polyphenylene ether-polyamide compositions having unexpectedly improved paint adhesion may be prepared by incorporating at least one paint adhesion improver selected from the class of polymeric materials consisting of

- a) homopolymers or copolymers of unsaturated nitrile compound, and
- b) alkyl acrylate or methacrylate homopolymers or copolymers.

In general the compositions of the present invention are prepared from

- (A) at least one polyphenylene ether and
- (B) at least one polyamide and

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- (C) at least one paint adhesion improver selected from the class of polymeric materials consisting of
  - a) homopolymers or copolymers of unsaturated nitrile compounds and
  - b) alkyl(meth)acrylate homopolymers and copolymers and
- (D) at least one compatibilizing agent.

Especially preferred compatibilizing agents will be selected from the group consisting of

- (a) liquid diene polymers
- (b) epoxy compounds
- (c) unsaturated functional compounds having in the molecule both (i) at least one carbon-carbon double bond or carbon-carbon triple bond and (ii) at least one carboxylic acid, acid halide, anhydride, acid anhydride, acid amide, acid ester, imido, amino or hydroxyl group
- (d) aliphatic polycarboxylic acid compounds or the derivatives thereof having both (i) a group represented by the formula-(OR) wherein R is hydrogen or an alkyl, aryl, acyl or carbonyl dioxy group and (ii) at least two groups, each of which may be the same or different, selected from carboxylic acid, acid halide, acid anhydride, anhydride, acid anhydride halide, acid ester, acid amide, imido, amino and salts thereof
- (e) polyfunctional acid halide compounds having both (i) an acid halide group and (ii) a group selected from carboxylic acid, carboxylic acid anhydride, acid ester or acid amide

(E) optionally one or more of rubbery polymers or copolymers as an impact strength improver.

The composition of the present invention will generally be prepared from 5 to 95, preferably 30 to 70 percent by weight polyphenylene ether (A), and from about 5 to 95, preferably 30 to 70 percent by weight polyamide (B), 0.5 to 50, preferably 1 to 20 parts by weight of at least one of the paint adhesion improvers (C), based on 100 by weight of the total of the polyphenylene ether and polyamides, and, 0.01 to 30, preferably from about 0.1 to about 5 parts by weight of at least one compatibilizer based on 100 by weight of the total of the polyphenylene ether and polyamides.

When employed, the impact strenth improver will generally be used in an amount of from about 2 to about 100 parts, preferably from about 5 to about 50 parts by weight based on 100 parts by weight of the total of the polyphenylene ether and polyamides.

# **DETAILED DESCRIPTION**

The polyphenylene ether used in the present invention is homopolymer or copolymer composed of the following repeating unit (I) or (I) and (II):

wherein  $R_1$ ,  $R_2$ ,  $R_3$ ,  $R_4$ ,  $R_5$  and  $R_6$  which may be identical or different each represents a monovalent residue such as an alkyl group of 1 - 4 carbon atoms excluding tert-butyl group, an aryl group, a halogen atom or a hydrogen atom, and  $R_3$  and  $R_5$  cannot be simultaneously hydrogen atom.

The polyphenylene ether may be a mixture of said homopolymer and said copolymer, or a graft copolymer of said polymer with styrene.

The homopolymer of polyphenylene ether includes poly(2,6-diethyl-1,4-phenylene)ether, poly(2-methyl-6-ethyl-1,4-phenylene)ether, poly(2,6-diethyl-1,4-phenylene)ether, poly(2-ethyl-6-n-propyl-1,4-phenylene)ether, poly(2,6-di-n-propyl-1,4-phenylene)ether, poly(2-methyl-6-n-butyl-1,4-phenylene)ether, poly(2-ethyl-6-isopropyl-1,4-phenylene)ether, poly(2-methyl-6-chloro-1,4-phenylene)ether, poly(2-methyl-6-hydroxyethyl-1,4-phenylene)ether and poly(2-methyl-6-chloroethyl-1,4-phenylene)ether.

The copolymer of polyphenylene ether includes polyphenylene ether copolymers mainly composed of polyphenylene ether structure which is obtained by copolymerization with o-cresol or an alkyl-substituted phenol such as 2,3,6-trimethylphenol which is represented by the formula (III):

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wherein R<sub>3</sub>, R<sub>4</sub>, R<sub>5</sub> and R<sub>6</sub> each represents a monovalent residue such as an alkyl group of 1 - 4 carbon atoms excluding tert-butyl group, an aryl group, a halogen atom or a hydrogen atom, and R3 and R5 cannot be simultaneously hydrogen atom.

The polyamide component (B) used in the present is well known in the art and may be selected from any of alliphatic polyamides or thermoplastic aromatic copolyamides or a combination thereof. The aliphatic polyamides have a molecular weight of 10,000 or more and can be produced by bonding of equimolar of a saturated aliphatic dicarboxylic acid of 4 - 12 carbon atoms and an aliphatic diamine of 2 - 12 carbon atoms. However, in the production, if necessary, the diamines may be excessively used so as to provide more amine terminal groups than carboxyl terminal groups in the polyamide, or alternatively, a dibasic acid may be excessively used so as to provide more acid groups. Similarly, these polyamides can be conveniently produced from acid production derivatives and amine production derivatives such as esters, acid chlorides and amine salts, of the above mentioned acids and amines.

Typical examples of the aliphatic dicarboxylic acid used for the production of the polyamides include adipic acid, pimelic acid, azelaic acid, suberic acid, sebacic acid and dodecanedioic acid.

On the other hand, typical examples of the aliphatic diamines include hexamethylenediamine and octamethylenediamine, and the like.

In addition, the aliphatic polyamides may also be produced by self-condensation of lactam. Examples of the aliphatic polyamides are polyhexamethylene adipamide (nylon 66), polyhexamethylene azelamide (nylon 69), polyhexamethylene sebacamide (nylon 610), polyhexamethylene dodecanamide (nylon 612), poly-bis-(p-aminocyclohexyl)methane dodecanoamide, polytetramethylene adipamide (nylon 46) and polyamides produced by ring cleavage of lactam such as polycaprolactam (nylon 6) and polylauryl lactam. Furthermore, there may be used polyamides produced by polymerization of at least two amines or acids selected from those used for the production of the above-mentioned polymers, for example, polymers produced from adipic acid, sebacic acid and hexamethylenediamine. The aliphatic polyamides futher include blends of above-mentioned polyamides such as a blend of nylon 6 and nylon 66 including copolymers such as nylon 66/6.

Preferably, the aliphatic polyamides used in the present invention are polyhexamethylene adipamide (nylon 66), polycaprolactam (nylon 6) and a blend of polyhexamethylene adipamide (nylon 66) with polycaprolactam (nylon 6).

The thermoplastic aromatic copolyamide is a copolyamide containing an aromatic component therein, for example, polyhexamethylene isophthalamide (nylon 6l). The copolyamide containing an aromatic component therein means a melt-polymerizable polyamide containing as a main component an aromatic amino acid and/or an aromatic dicarboxylic acid such as para-aminomethylbenzoic acid, para-aminoethylbenzoic acid, terephthalic acid and isophthalic acid. Diamines which may constitute another component of the polyamide include hexamethylenediamine, undecamethylenediamine, dodecamethylenediamine, 2,2,4-/2,4,4-trimethylhexamethylenediamine, m-xylylenediamine, p-xylylenediamine, bis(p-aminocyclohexyl)methane, bis(p-aminocyclohexyl)propane, bis(3-methyl-4-aminocyclohexyl) methane, 1,3-bis(aminomethyl)cyclohexane and 1,4-bis(aminomethyl)cyclohexane. An isocyanate may also be used in place of the diamine.

Any other comonomers may be used, if necessary. Examples of the comonomers are 4,4'-diphenyl-methane diisocyanate and tolylene diisocyanate. Other examples thereof are a unit of lactam, a unit of ω-amino acid of 4 - 12 carbon atoms, a compound derived from an aliphatic dicarboxylic acid of 4 - 12 carbon atoms and an aliphatic diamines of 2 - 12 carbon atoms, for example, lactams and amino acids such as ε-caprolactam, ω-caprolactam, 11-aminoundecanoic acid and 12-aminododecanoic acid, and equimolar salts of the above-mentioned various diamines and adipic acid, azelaic acid or sebacic acid.

Typical examples of the thermoplastic aromatic copolyamides comprising these components are copolymer polyamide of p-aminomethylbenzoic acid and ε-caprolactam (nylon AMBA/6), polyamides mainly composed of 2,2,4-/2,4,4-trimethylhexamethylenediamine \*terephthalate (nylon TMDT and nylon TMDT/6I), hexamethylenediamine isophthalate mainly composed of polyamides bis(p-aminocyclohexyl)hexamethylenediamine\*terephthalate and containing, as a comonomer, methane\*terephthalate, and/or bis(3-methyl-4-aminocyclohexyl)methane\*isophthalate and/or bis(3-methyl-4aminocyclohexyl)propane\*isophthalate and/or bis(p-aminocyclohexyl)propane\*terephthalate (nylon 6l/PACm I, nylon 6I/ DMPACM I, nylon 6I/PACP I, nylon 6I/6T/PACM I/PACm T, nylon 6I/6T/DMPACM I/DMPACM T, nylon 61/6T/PACP I/PACP T), polyamides mainly composed of hexamethylenediamine isophthalate or hexamethylenediamineterephthalate and containing, as a comonomer, ¿-caprolactam, 12-aminododecanoic bis(p-aminocyclohexyl)methane adipate or hexamethylenediamine adipate, aminocyclohexly)methane adipate (nylon 61, 61/6T, 61/12, 6T/6, 6T/66, 6I/PACM 6, 6I/DMPACM 6), and polyamides mainly composed of bis(p-aminocyclohexyl)methane isophthalate or bis(3-methyl,4comonomer, as containing, а aminocyclohexyl)methane-isophthalate and hexamethylenediamine dodecanedioate or 12-amino-dodecanoic acid (nylon PACM I/612 and nylon DMPACM 1/12).

The aromatic nuclear-hydrogenated copolyamide of component (B) is an alicyclic copolyamide obtained by using cyclohexane 1,4-dicarboxylic aicd or cyclohexane 1,3-dicarboxylic acid obtained by nuclear-hydrogenation of terephthalic acid or isophthalic acid in place of terephthalic acid or isophthalic acid which is an acid component of the above-metioned aromatic copolyamides. Furthermore, nuclearhydrogenation product of diamines or diisocyanates such as 4,4 diphenylmethane diisocyanate or tolylene diisocyanate, may also be used as a monomer.

The paint adhesion improver (C) used for the practice of the present invention will be selected from the class of polymeric materials consisting of

a) homopolymers or copolymers of unsaturated nitrile compounds and

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b) alkyl acrylate or methacrylate homopolymers or copolymers as hereinafter defined.

The homopolymers or copolymers of unsaturated nitrile compounds a) useful for the practice of the present invention are well known in the art.

The term "unsaturated nitrile compound" used herein means a compound having in its moleculer structure at least one ethylenic carbon-carbon double bond or carbon-carbon triple bond and -C=N radical. The preferred unsaturated nitrile compounds are acrylonitrile and methacrylonitrile. The most preferred unsaturated nitrile compound is acrylonitrile. The illustrative examples of the homopolymer of the unsaturated nitrile compounds are polyacrylonitrile (PAN) and polymethacrylonitrile.

The illustrative examples of the copolymer of the unsaturated nitrile compound are styrene acrylonitrile random copolymer, random copolymer of acrylonitrile and butadiene, often called NBR, random copolymer of acrylonitrile and alkyl acrylate or methacrylate including but not limited to acrylonitrile/ethylacrylate copolymer, acrylonitrile/butylacrylate copolymer, acrylonitrile/methylacrylate/butyl acrylate terpolymer, acrylonitrile/styrene/ethylacrylate terpolymer, graft copolymer of polybutadiene and acrylonitrile and styrene, often called ABS resin, and the like. Content of the unsaturated nitrile compound in the homopolymer or copolymer of the unsaturated nitrile compound is desirably 20 - 80 % by weight. If it is less than 20 % by weight, paint adhesion is not sufficiently improved, and if more than 80 % by weight, much discoloration occurs after thermal aging. The content of the unsaturated nitrile compound is more desirably 20 - 55 % by weight. Obviously, copolymers or terpolymers of an unsaturated nitrile compound and an alkyl acrylate or methacrylate may be understood as either copolymers of unsaturated nitrile compounds or copolymers of alkyl acrylates or methacrylates.

The alkyl acrylate or methacrylate homopolymers or copolymers b) useful for the practice of the present invention are well known in the art. It is desirable that these alkyl acrylate or methacrylate homopolymers or copolymers are in glassy state at room temperature, or have a modulus of elasticity of about 10,000 kg/cm<sup>2</sup>. If these alkyl acrylate or methacrylate copolymers are in rubbery state at room temperature, it should be noted that copolymers or terpolymers of ethylene and alkyl acrylate or methacrylate having about 50 % by weight or higher ethylene are excluded from the paint adhesion improver as defined herein, since they exhibit lesser paint adhesion strength. The illustrative examples of the alkyl acrylate or methacrylate homopolymers or copolymers include polymethylmethacrylate, methylmetacylate/styrene copolymer, graft copolymer of styrene/methylmethacrylate/polybutadiene, often called MBS, methylmethacrylate/methylacrylate copolymer, methylmethacrylate/ethylacrylate copolymer, methylmethacrylate/butylacrylate copolymer, terpolymer, ethylacrylate/butylacrylate/methoxyacrylate methylmethacrylate/glycidylmethacrylate ethylacrylate/butylacrylate copolymer, styrene/methylmethacrylate/glycidylmethacrylate terpolymer, copolymers of alkylacrylates and at least one

crosslinkable comonomer, often called acryl rubbers, and the like. The above mentioned paint adhesion improver may be employed alone or in any combination of one another.

Examples of preferred compatibilizing agents (D) that may be employed in the practice of the present invention are

- (a) liquid diene polymers,
- (b) epoxy compounds,

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- (c) unsaturated functional compounds,
- (d) aliphatic polycarboxylic acid compounds or the derivatives thereof as describled hereunder, and
- (e) polyfunctional acid halide compounds as described hereunder.

Liquid diene polymers(a) suitable for use herein include homopolymers of a conjugated diene and copolymers of a conjugated diene with at least one monomer selected from the group consisting of other conjugated dienes, olefins, e.g. ethylene, propylene, butene-1, isobutylene, hexene-1, octene-1 and dodecene-1, and mixtures thereof. The polymers(a) have a number average molecular weight of from 150 to 10,000,preferably 150 to 5,000. These homopolymers and copolymers include, among others, polybutadiene, polyisoprene, poly(1,3-pentadiene), poly(butadiene-isoprene), poly(styrene-butadiene), polychloroprene, poly(butadiene-alpha methyl styrene), poly(butadiene-styrene-isoprene), poly(butylene-butadiene) and the like.

Epoxy compounds (b) suitable for use in the practice of the present invention there are given (1) epoxy resins produced by condensing polyhydric phenols (e.g. bisphenol-A, tetrabromobisphenol-A, resorcinol and hydroquinone) and epichlorohydrin; (2) epoxy resins produced by condensing polyhydric alcohols (e.g. ethylene glycol, propylene glycol, butylene glycol, polyethylene glycol, polypropylene glycol, pentaerythritol and trimethylolethane and the like) and epichlorohydrin; (3) glycidyletherified products of monohydric compounds including phenyl glycidylether, allyl glycidylether, butyl glycidylether and cresyl glycidylether; (4) glycidyl derivatives of amino compounds for example, the diglycidyl derivative of aniline, and (5) epoxidized products of higher olefinic or cycloalkene, or natural unsaturated oils (e.g. soybean) as well as of the foregoing liquid diene polymers.

The unsaturated functional compounds (c) are those having in the molecule both (i) an ethylenic carbon-carbon double bond or a carbon-carbon triple bond and (ii) at least one carboxylic acid, acid anhydride, acid halide, anhydride, acid halide anhydride, acid amide, acid ester, imide, amino, or hydroxy group. Examples of such unsaturated polyfunctional compounds are maleic acid; maleic anhydride; fumaric acid; citraconic acid; itaconic acid; maleimide; maleic hydrazine; reaction products resulting from a diamine and maleic anhydride, maleic acid, fumaric acid, etc.; dichloromaleic anhydride; maleic acidamide; unsaturated monocarboxylic acid (such as acrylic acid, butenoic acid, methacrylic acid, α-ethylacrylic acid, pentenoic acid, decenoic acids, undecenoic acids, butenoic acids, linoleic acid,); esters, acid amides or anhydrides of the foregoing unsaturated carboxylic acids including glycidyl(meth)acrylate; unsaturated alcohols (such as allyl alcohol, crotyl alcohol, methyl vinyl carbonol, 4-pentene-1-ol, 1,4-hexadiene-3-ol, 3-butene-1,4-diol, 2,5-dimethyl-3-hexene-2,5-diol and alcohols of the formula C<sub>n</sub>H<sub>2n-5</sub>OH, C<sub>n</sub>H<sub>2n-7</sub>OH and C<sub>n</sub>H<sub>2n-9</sub>OH, wherein n is a positive integer up to 30), unsaturated amines resulting from replacing the -OH group(s) of the above unsaturated alcohols with NH<sub>2</sub> groups; and functionalized diene polymers and copolymers.

The aliphatic polycarboxylic acid compounds or the derivatives thereof (d) are represented by the formula

(RIO)mR(COORII)n(CONRIIIRIV)

wherein R is a linear or branched chain, saturated aliphatic hydrocarbon of from 2 to 20, preferably 2 to 10 carbon atoms; R<sup>I</sup> is selected from the group consisting of hydrogen or an alkyl, aryl, acyl or carbonyl dioxy group of 1 to 10, preferably 1 to 6, most preferably 1 to 4 carbon atoms, especially preferred is hydrogen; each R<sup>II</sup> is independently selected from the group consisting of hydrogen or an alkyl or aryl group of from 1 to 20 carbon atoms, preferably from 1 to 10 carbon atoms; each R<sup>III</sup> and R<sup>IV</sup> is independently selected from the group consisting essentially of hydrogen or an alkyl or aryl group of from 1 to 10, preferably from 1 to 6, most preferably 1 to 4 carbon atoms; m is equal to 1 and (n + s) is greater than or equal to 2, preferably equal to 2 or 3, and n and s are each greater than or equal to zero and wherein (OR<sup>I</sup>) is alpha or beta to a carbonyl group and at least two carbonyl groups are separated by 2 to 6 carbon atoms. Obviously, R<sup>I</sup>, R<sup>II</sup>, R<sup>III</sup> and R<sup>IV</sup> cannot be aryl when the respective substituent has less than 6 carbon atoms.

Illustrative examples of suitable polycarboxylic acids are citric acid, malic acid, and agaricic acid.

The polyfunctional acid halide compounds (e) suitable for use herein are characterized as having both (i) at least one acid halide group, preferably acid chloride group and (ii) at least one carboxylic acid, carboxylic acid anhydride, acid ester or acid amide group, preferably a carboxylic acid or carboxylic acid anhydride group. Examples of compatibilizers within this group are trimellitic anhydride acid chloride,

chloroformyl succinic anhydride, and the like.

Examples of impact strength improver (E) that may be employed in the practice of the present invention are,

a polyolefins,

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- (bethylene/ olefin copolymer rubbers,
- ©ethylene/ olefin/polyene terpolymer rubbers,
- (d) functionalized aforementioned olefin polymers by grafting at least one unsaturated functional compound thereto,
  - @copolymers or terpolymers of ethylene and at least one unsaturated functional compound,
- (f)diene rubbers, and
  - hydrogenated or nonhydrogenated block copolymers of vinyl-aromatic compound and diene.

Polyolefins (a) suitable for use in the practice of the present invention include high density polyethylene, low density polyethylene, linear low density polyethylene, polypropylene, crystalline propylene/ethylene copolymer, polybutene-1, polymethylpenten-1, polyisobutylene and the like. Ethylene/olefin copolymer rubbers (b) for use in the practice of the present invention include ethylene/propylene copolymer rubber, often called EPR, ethylene/butene-1 copolymer rubber and the like. Ethylene/olefin/polyene terpolymer rubbers © for use in the practice of the present invention include ethylene/propylene/ethylidenenorbornen terpolymer rubber, ethylene/propylene/dicyclopentadiene terpolymer rubber, ethylene/propylene/1,4 hexadiene terpolymer rubber and the like, often called EPDM. Functionalized olefin polymers (d) useful for the practice of the present invention include olefin polymers described in the above a , b and c with at least one unsaturated functional compound grafted thereto. The illustrative examples of the unsaturated functional compounds are acrylic acid, methacrylic acid, alkylester derivatives thereof, such as methyl acrylate or methacrylate, ethyl acrylate or methacrylate, butyl acrylate or methacrylate and the like, dicarboxylic acid or acid anhydride such as fumaric acid, maleic acid, 25 maleic anhydride, itaconic acid and the like, acrylamide, N-(hydroxymethyl)acrylamide, glylidyl derivatives of acrylic or methacrylic acid such as glycidyl acrylate or methacrylate, vinyl acetate, vinyl chloride, styrene and the like. Method for the grafting of the unsaturated functional compound to the olefin polymers is not critical in the practice of the present invention and any known method in the art may be employed. Melt mixing of the olefin polymers and the unsaturated functional compounds with a suitable amount of a free radical initiator may be employed. Grafting of the unsaturated functional compound under an aqueous suspension of olefin polymers with a suitable amount of a free radical initiator and a dispersing agent may also be employed. Copolymers or terpolymers (e) of ethylene and at least one unsaturated functional compound useful for the practice of the present invention include ethylene/acrylic or methacrylic acid copolymer, ethylene/alkyl acrylate or methacrylate copolymer, ethylene/vinylacetate copolymer, ethylene/alkyl acrylate or methacrylate/maleic anhydride terpolymer, ethylene/methyl acrylate or methacrylate/glycidyl acrylate or methacrylate terpolymer and the like. Diene rubbers ① useful for the practice of the present invention include polybutadiene, styrene butadiene random copolymer, often called SBR, natural rubber, polyisoprene, and the like.

Hydrogenated or nonhydrogenated block copolymers @ of vinyl aromatic compound and diene are well known in the art. Illustrative examples of the block copolymers are styrene/butadiene diblock copolymer, styrene/isoprene diblock copolymer, styrene/butadiene/styrene triblock copolymer, styrene/isoprene/styrene triblock copolymer, radial teleblock copolymers of styrene and butadiene, hydrogenated products of the foregoing block copolymers and the like. The foregoing impact strength improver (E) may be used alone or in any combination of one another.

The total amount of one or more of the compatibilizing agent(D) used herein will be dependent on the specific compatibilizing agent selected. It is desirable to use at least necessary enough amount in enhancing the compatibility of the polyphenylene ether/polyamide blend. Generally the amount of compatibilizing agent will be from about 0.01 to 30, preferably from about 0.1 to about 5 parts by weight per 100 parts by weight of the polyphenylene ether/polyamide blend.

The total amount of one or more of the paint adhesion improver(C) used herein will be dependent on the specific paint adhesion improver selected and the specific polymer blend system in question. It is desirable to use at least necessary enough amount in improving the polyphenylene ether/polyamide blend. Generally the amount of the paint adhesion improver will be from about 0.5 to 40, preferably 1 to 20 parts by weight per 100 parts by weight of the polyphenylene ether/polyamide blend.

Blending method of the components (A), (B), (C), (D), and if employed, (E) is not critical. Known melt kneading methods can be employed as the blending method. Extruders, kneaders, rolls and the like may be used. Preferably extruders can be used as melt kneading apparatuses. There is no special limitation in sequence or addition of the components upon melt kneading. That is, there are a method which comprises

adding all of the components (A), (B), (C) and (D), at the same time and simultaneously melt-kneading them and a method which comprises pre-meltkneading components (A), (C), and (D) in the presence or absence of a free-radical initiator and then adding component (B) and melt-kneading them. When component (E) as an impact strength improver is added, there may be also the following methods: a method which comprises adding component (E) together with components (A), (B), (C), and (D) at the same time and simultaneously melt-kneading them; a method which comprises pre-meltkneading component (E) with components (A), (C) and (D) in the presence or absence of a free-radical initiator and then adding component (B), and melt-kneading them. The foregoing polyphenylene ether/polyamide blends may further comprise inorganic fillers such as talc, aluminosilicate, mica, carbon black, glass fiber and the like, pigments, heat stabilizers, ultraviolet degradation inhibitors, antioxidants, flame retardants, plasticizers and the like.

The molded products obtained from the thermoplastic resin composition of the present invention can be used suitably for automobile parts and electrical and electronic parts.

As examples of automobile parts to which the composition of the present invention can be applied, mention may be made of exterior trim parts such as bumper, fender, apron, hood panel, fascia, rocker panel, rocker panel reinforce, floor panel, rear quarter panel, door panel, door support, roof top, and trunk lid, interior trim parts such as instrument panel, console box, glove box, shift knob, pillar garnish, door trim, handle, arm rest, wind louver, carpet, seat belt, and seat, interior parts of engine room such as distributor cap, air cleaner, radiator tank, battery case, radiator shroud, washer tank, cooling fan, and heater case, mirror body, wheel cover, trunk trim, trunk mat and gasoline tank.

The following examples further illustrate the present invention, but the present invention is not limited to them. In the preparation of the polyphenylene ether/polyamide blends disclosed in the examples herein, a twin screw extruder TEX44 made by Nippon Seikosho Kabushiki Kaisha was used, wherein L/D ratio of the extruder is 32 and the first and the second openings are provided at the positions of L/D ratios of 1 and 16, respectively, (L: the length of the screw, D: the diameter of the screw). The cylinder temperature was set at about 260° C and screw speed was set at 360 rpm.

The formulations of the individual blends were shown in Table 1. The paint adhesion strength measured of each blend was also shown in Table 1.

In Table 1, compatibilizers and free radical initiators were shown in parts by weight per 100 parts of the total of the polymeric materials and each of the polymeric materials was shown in weight % of the total of the polymeric materials.

The ingredients listed in the column "Feed-1" of Table-1 were fed from the first feed opening. The ingredients listed in the column "Feed-2" of Table-1 were fed from the second feed opening. All ingredients fed together either from the first feed opening or the second feed opening were mixed well by a tumbler mixer prior to the feeding.

The feed rate of the first feed and the second feed were controlled by the automatic weight feed control system so as to maintain the formulation of the idividual blend as specified in Table-1.

Polyphenylene ethers employed in the examples:

polyphenylene ether: a reduced viscosity of 0.52 dl/g measured at 25 °C in a chloroform solution of 0.5 g/dl concentration manufactured by Nippon Polyether Yugen Kaisha (hereinafter denoted as PPE-A);

polyphenylene ether having a reduced viscosity of 0.42 dl/g under the same conditions as above (hereinafter denoted as PPE-B).

Polyamides:

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Nylon 6 (Unitika Kabushiki Kaisha's Nylon 6 (A1030 BRL));

Nylon 66 (UBE Kosan Kabushiki Kaisha's UBE nylon 66 (2015B));

Aromatic polyamides:

EMS Chemie's Grivory @ XE3038;

Huels AG's Trogamid ® T 2010.

Free radical initiators:

dicumyl peroxide (Sanperox ® DCP made by Sanken Kako Kabushiki Kaisha)

The free radical initiator was always preblended with polyphenylene ether and fed from the first feed opening. Polyphenylene ether was always fed from the first feed opening and polyamides were always fed from the second opening.

The paint adhesion improver (C) used in the examples were as follows;

C1) Acrylonitrile styrene copolymer (SAN)

SAN-1 (weight average molecular weight: 140,000, acrylonitrile content: 23 weight %);

SAN-2 (weight average molecular weight: 120,000, acrylonitrile content: 27 weight %);

SAN-3 (weight average molecular weight 170,000, acrylonitrile content: 27 weight %).

C2) Acrylonitrile and styrene-grafted polybutadiene (ABS)

ABS-A: about 50 weight % of polybutadiene and about 15 weight % of acrylonitrile;

ABS-B: about 30 weight % of polybutadiene and about 20 weight % of acrylonitrile.

C3) Acryl rubber (AR)

Nippon Zeon Kabushiki Kaisha's Nipol ® AR-32 (hereinafter denoted as AR). Mooney viscosity: about 40 (at 100 °C).

C4) Acrylonitrile/butadiene rubber (NBR) Nippon Zeon Kabushiki Kaisha's Nipol @.

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|       |                | Acrylonytrile content |
|-------|----------------|-----------------------|
| NBR-A | Nipol® DN-002  | 53 wt %               |
| NBR-B | Nipol® DN-115  | 42 wt %               |
| NBR-C | Nipol® DN 402  | 23 wt %               |
| NBR-D | Nipol® DN 401L | 18 wt %               |

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C5) Polymethylmethacrylate (PMMA)

Sumitomo Chemical Company, Ltd.'s Sumipex ® LG,

C6) Glycidylmethacrylate/methylmethacrylate/ethylacrylate terpolymer (hereinafter denoted as MEG). Negami Kogyo Kabushiki Kaisha's High Pearl ® MG-10 where glycidylmethacrylate: about 10 weight %, methylmethacrylate: about 45 weight %, ethylacrylate: about 45 weight %. Molecular weight: about 30,000.

C7) Pólyacrylonitrile (PAN)

A copolymer of acrylonitrile (about 90 weight %) and methylacrylate (about 10 weight %). Molecular weight: about 100,000 (hereinafter denoted as PAN).

The impact strength improver (E) used in the examples were as follows;

E1) Acrylamide-grafted ethylene/propylene copolymer rubber (EPR) (denoted hereinafter as mEPR-A);

maleated EPR (denoted as mEPR-B).

They were prepared by first mixing 100 parts by weight of EPR (Esprene ® E-120p), 0.1 part by weight of dicumylperoxide (DCP) as a free radical initiator and either 1.5 parts by weight of acrylamide or maleic anhydride (MAH) and then melt-extruding the premix at a temperature of about 230 °C utilizing the TEX44 twin screw extruder and pelletized, cooled and dried.

E2) Styrene grafted EPR (denoted hereinafter as Sg-EPR)

Sg-EPR was prepared by the graft polymerization in solution. Sg-EPR was obtained containing about 20 weight % of polystyrene.

E3) Acrylamide-grafted styrene-grafted EPR (denoted hereinafter as m-Sg-EPR)

It was prepared by graft polymerization in melting of 100 parts by weight of the Sg-EPR and 1.5 parts by weight of acrylamide.

E4) Styrene-grafted EPDM (denoted hereinafter as Sg-EPDM)

It was prepared by the graft polymerization in solution. Thus Sg-EPDM was obtained containing about 29 weight % of polystyrene.

E5) Maleic anhydride-grafted styrene-grafted EPDM (denoted hereinafter as mSg-EPDM)

It was prepared by graft polymerization in melting of 100 parts by weight of Sg-EPDM and 1.5 parts by weight of maleic anhydride.

E6) N-(hydroxymethyl)acrylamide-grafted ethylene/butene-1 copolymer rubber (hereinafter denoted as mEBR)

It was prepared by graft polymerization in melting of 100 parts by weight of ethylene/butene-1 copolymer rubber (EBR, Sumitomo Chemical Co., Ltd., about 30 weight % of butene-1 and the Mooney viscosity measured at 120 °C: about 20) and 1.5 parts by weight of N-(hydroxymethyl)acrylamide.

E7) Styrene-grafted ethylene/butene-1 copolymer rubber (denoted hereinafter as Sg-EBR).

It was prepared by substituting the EPR used in the preparation of Sg-EPR with the same EBR as used in the preparation of mEBR.

E8) Shell Kagaku's Kraton D1101

Styrene butadiene triblock copolymer (denoted hereinafter as SBS).

E9) Shell Kagaku's Kraton G1701

A hydrogenated styrene/isoprene diblock copolymer (denoted hereinafter as SEP).

E10) Norsolor's Lotader 4700

An ethylene/ethylacrylate/maleic anhydride terpolymer (denoted hereinafter as LOT).

- E11) Sumitomo Chemical Co., Ltd.'s glycidylmethacrylate/methylacrylate/ethylene terpolymer having 3 weight % of glycidylmethacrylate, 30 weight % of methyl acrylate and the balance being ethylene, a terpolymer of ethylene, glycidylmethacrylate and methylacrylate (hereinafter denoted as EGMA).
  - E12) Polystyrene

Nippon Polystyrene Kabushiki Kaisha's Esbrite ® T8K; Esbrite ® 500HR-Y3 (high impact polystyrene denoted as HIPS hereinafter).

E13) N-(hydroxymethyl)acrylamide-grafted ethylene/propylene/diene terpolymer rubber (denoted hereinafter as m-EPDM-A)

It was prepared by substituting the EBR used in the preparation of mEBR with the EPDM.

The paint adhesion test was performed in the following manner;

- 1) The granules of each of the blends prepared according to the formulation listed in Table 1 to be tested were always dried at 100°C for 6 hours with preheated dry air stream, the dew point of which was about -40°C, and cooled thereafter by room temperature dry air stream.
- 2) The blend granules were injection molded to form test plates of 150 milimeter x 150 milimeter x 3 milimeter size at a temperature range of about 290 °C.
- 3) Paint spraying was made with a paint spray gun onto the resulting plates to form a paint layer of about 25 micron thick.
  - 4) The paint-coated plates were then cured for 30 minutes at 70 °C in an infrared paint cure oven.
- 5) The paint layer peeling test was performed at 1 hr, 2 hr, 5 hr, 12 hr and 24 hr after the paint curing.

The cured paint layer was cross cut by a sharp edged knife so as to make one hundred pieces of 2 mm x 2 mm square each. Then a sheet of adhesive tape was applied to the cross cut surface and the tape was peeled off.

The paint adhesion strength was measured by counting the number of about 2 mm by 2 mm square pieces remained unpeeled off, out of the 100 cross cut pieces.

6) The paint used in the examples was "Origiplate Z-NY metalic silver" manufactured by Origin Denki Kabushiki Kaisha.

The heat discoloration test was performed in the following manner:

- 1) Plates of 150 mm x 150 mm x 3 mm (thickness) were placed in a Geer's oven at 160°C for 1 hour, and then were took out to evaluate degree of discoloration.
  - 2) Evaluation:

Degree of discoloration

X,  $\triangle$ ,  $\bigcirc$  (Great) (Little)

Example-1

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15 Kg of polyphenylene ether (PPE-A), 0.25 Kg of maleic anhydride (MAH), 5 grams of dicumylperoxide (DCP), and 10 Kg of ABS-A were premixed as prescribed in the Feed-1 column of Example-1 of Table-1, using a 100 liter tumbler mixer blending the ingredients for 3 minutes prior to the feeding to the first feed opening of the TEX-44 twin screw extruder.

The premix prepared for the first feed opening will be referred to as the first feed premix hereinafter. 20 Kg of the nylon 6 and 5 Kg of the nylon 66 were premixed as prescribed in the Feed-2 column of Example-1 of Table-1 using the same tumbler mixer for 3 minutes prior to the feeding to the second feed opening of the TEX-44 twin screw extruder.

The premix prepared for the second feed opening will be referred to as the second feed premix hereinafter. The cylinder temperature of the extruder was set at about 260 °C and the screw speed, at about 360 rpm. The first feed premix was fed to the first feed opening at a rate of 20.2 Kg/Hr and the second feed premix was fed to the second feed opening at a rate of 20 Kg/Hr so as to maintain the formulation of the

ingredients shown in Example-1.

The resulting blend was cooled in a water bath by chilled water of about 10 °C and pelletized and dried. Thus prepared blend granules were injection molded to make 5 plates of 150 mm x 150 mm x 3 mm test specimen.

The paint adhesion strength was tested with 5 of the plates according to the test method described before. The average of the paint adhesion strength measured of the five plates was shown in the table 1.

#### Example-2

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The experiment was conducted in the same manner as in Example-1 except that ABS-A was substituted with SAN-1.

#### 15 Comparative example-1

The experiment was conducted in the same manner as in Example-1 except that ABS-A was substituted with polystyrene (T8K).

20 Examples 3 through 25 and Comparative Examples 2 through 9.

The experiments were conducted in the same manner as in Example-1 except that

- (1) About 20.25 kg each of the first feed premix was prepared according to the formulation shown in Feed-1 column of each of the examples.
- (2) 20 Kg each of the second feed premix was prepared according to the formulation shown in Feed-2 column of each of the examples.

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Table 1

|    |        |                          | <del></del>  |             |                             | ı — - 1                |                        |                        |
|----|--------|--------------------------|--------------|-------------|-----------------------------|------------------------|------------------------|------------------------|
| 5  |        |                          | Example (EX) | EX<br>2     | Compar-<br>ative<br>EX<br>1 | EX<br>3                | EX<br>4                | EX<br>5                |
| 10 |        | PPE-A                    | 30           | 30          | 30                          | -                      | -                      | -                      |
|    |        | PPE-B                    | -            | -           | -                           | 40                     | 40                     | 40                     |
| 15 |        | Compatibilizer           | MAH<br>0.5   | MAH<br>0.5  |                             | Fumaric<br>acid<br>0.5 | Fumaric<br>acid<br>0.5 | Fumaric<br>acid<br>0.5 |
|    | Feed-1 | Free radical initiator   | 0.01         | 0.01        | 0.01                        | -                      | -                      | -                      |
| 20 |        | Paint adhesion improver  | ABS-A<br>20  | SAN-1<br>20 | _                           | ABS-B<br>5             | NBR-A<br>5             | PMMA<br>5              |
|    |        | Impact strength improver | -            | -           | -                           | πEPR-A<br>5            | mEPR-A<br>5            | mEPR-A<br>5            |
| 25 |        | Polystyrene              | _            | _           | T8K<br>20                   | -                      | -                      | -                      |
| 30 |        | Nylon 6                  | 40           | 40          | 40                          | 40                     | 40                     | 40                     |
| -  |        | Nylon 66                 | 10           | 10          | 10                          | 10                     | 10                     | 10                     |
| 35 |        | Aromatic<br>polyamide    | _            | -           | -                           | -                      | -                      | -                      |
|    | Feed-2 | Paint adhesion improver  | -            | -           | -                           | _                      | _                      | -                      |
| 40 |        | Impact strength improver | -            | _           | -                           | -                      | -                      | -                      |
|    |        | Polystyrene              | _            | _           | -                           | -                      | -                      |                        |
| 45 |        | Paint 1 hr               | 59           | 96          | 0                           | 0                      | 0                      | 0                      |
|    |        | adhesion 2 hr            | 62           | 97          | 0                           | 0                      | 0                      | 0                      |
|    |        | strength 5 hr            | 82           | 99.8        | 74                          | 21                     | 9                      | 11                     |
| 50 |        | 12 hr                    | 96           | 100         | 92                          | 58                     | 28                     | 27                     |
|    |        | 24 hr                    | 99.9         | 100         | 96                          | 90                     | 78                     | 79                     |
|    |        |                          |              |             |                             |                        |                        |                        |

(cont'd)

|           |        |                            |                        |                             | ·····      |              |                    |                    |
|-----------|--------|----------------------------|------------------------|-----------------------------|------------|--------------|--------------------|--------------------|
| 5         |        |                            |                        | Compar-<br>ative<br>EX<br>2 | EX<br>7    | EX<br>8      | EX<br>9            | EX<br>10           |
|           |        | PPE-A                      | _                      | -                           | 35         | 35           | 40                 | 40                 |
| 10        |        | PPE-B                      | 40                     | 40                          | -          | -            | -                  | -                  |
| 15        |        | Compatibilizer             | Fumario<br>Acid<br>0.5 | Fumaric<br>Acid<br>0.5      | МАН<br>0.5 | MAH<br>0.5   | MAH<br>0.5         | МАН<br>0.5         |
|           | Feed-1 | Free radical<br>initiator  | -                      | -                           | -          | -            | 0.1                | 0.1                |
| 20        |        | Paint adhesion<br>improver | ABS-A<br>3             | -                           | ABS-B<br>5 | NBR-A<br>5   | -                  | -                  |
|           |        | Impact strength improver   | mEPR-B                 | mEPR-B                      | Sg-EBR     | mSg-EF       | mSg-<br>EPDM<br>10 | mSg-<br>EPDM<br>10 |
| 25        |        | Polystyrene                | _                      | -                           | -          | -            | -                  | <b>-</b>           |
|           |        | Nylon 6                    | 40                     | 40                          | 45         | -            | 44                 | 45                 |
| 30        |        | Nylon 66                   | 10                     | 10                          | -          | -            | _                  | -                  |
|           |        | Aromatic<br>polyamide      | -                      | -                           | _ 6        | rivory<br>45 | Troga-<br>mid<br>1 | _                  |
| 35        | Feed-2 | Paint adhesion improver    | NBR-A<br>2             | -                           |            | _            | NBR-A<br>5         | PMMA<br>5          |
|           |        | Impact strength            | _                      | -                           | -          | -            | _                  |                    |
| <b>40</b> |        | Polystyrene                | -                      | -                           | HIPS<br>5  | HIPS<br>5    | -                  | _                  |
| 45        |        | Paint 1 hr                 | 0                      | 0                           | 43         | 5            | 7                  | 9                  |
|           |        | adhesion 2 hr              | 0                      | 0                           | 58         | 42           | 49                 | 48                 |
|           |        | strength 5 hr              | 28                     | 0                           | 83         | 63           | 68                 | 73                 |
| 50        |        | 12 hr                      | 46                     | 12                          | 90         | 81           | 83                 | 86                 |
|           |        | 24 hr                      | 85                     | 25                          | 98         | 90           | 92                 | 94                 |
|           |        |                            |                        |                             |            |              | /                  | 1.3.               |

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|        |        | •                          | Compar-<br>ative<br>EX<br>3 |                     | Compar-<br>ative<br>EX<br>4 | EX<br>12   | EX<br>13     | Compara-<br>tive<br>EX<br>5 |
|--------|--------|----------------------------|-----------------------------|---------------------|-----------------------------|------------|--------------|-----------------------------|
| _      |        | PPE-A                      | 40                          | -                   | -                           | _          | -            | -                           |
| )      |        | PPE-B                      | -                           | 40                  | 40                          | 40         | 40           | 40                          |
|        |        | Compatibilizer             |                             |                     | Citric<br>acid<br>0.5       | ман<br>0.5 | MAH<br>0.5   | MAH<br>0.5                  |
| ;<br>F | 'eed-1 | Free radical initiator     | -                           | -                   | -                           | 0.01       | 0.01         | 0.01                        |
| )      |        | Paint adhesion improver    | -                           | _                   | -                           | NBR-A<br>5 | AR<br>5      | -                           |
|        |        | Impact strength improver   | mSg-<br>EPDM<br>10          | SEP<br>10           | SEP<br>10                   | mEBR<br>5  | mEBR<br>5    | mEBR<br>10                  |
| ;      |        | Polystyrene                |                             | -                   | _                           | -          | -            | -                           |
| _      |        | Nylon 6                    | 45                          | -                   | -                           | 45         | 45           | 45                          |
| )      |        | Nylon 66                   | -                           | -                   | -                           | -          | -            | -                           |
|        |        | Aromatic<br>polyamide      | -                           | Troga-<br>mid<br>45 | Troga-<br>mid<br>45         | -          | -            | _                           |
| 5<br>F | Feed-2 | Paint adhesion<br>improver | _                           | AR<br>5             | _                           | -          | _            | -                           |
| )      |        | Impact strength improver   | _                           | _                   | -                           | LOT<br>5   | EQMA<br>5    | LOT<br>5                    |
|        |        | Polystyrene                | HIPS<br>5                   | _                   | HIPS<br>5                   | _          | <del>-</del> | -                           |
| -<br>5 |        | Paint 1 hr                 | 0                           | 58                  | 0                           | 0          | 0            | 0                           |
|        |        | adhesion 2 hr              | 0                           | 61                  | 0                           | . 0        | 0            | 0                           |
|        |        | strength 5 hr              | 21                          | 85                  | 58                          | 8          | 23           | 0                           |
| 0      |        | 12 hr                      | 38                          | 92                  | 72                          | 29         | 47           | 0                           |
|        |        | 24 hr                      | 60                          | 99.1                | 89                          | 76         | 89           | 21                          |

| 5  |            |                          | EX<br>14    | EX<br>15           | Compar-<br>ative<br>EX<br>6 | EX<br>16     |             | Compara-<br>tive<br>EX<br>7 |
|----|------------|--------------------------|-------------|--------------------|-----------------------------|--------------|-------------|-----------------------------|
| 10 |            | PPE-A                    | -           | -                  | _                           | 40           | 40          | 40                          |
|    |            | PPE-B                    | 40          | 40                 | 40                          | -            | -           | -                           |
| 15 |            | Compatibilizer           | MAH<br>0.5  | MAH<br>0.5         | MAH<br>0.5                  | MAH<br>0.5   | MAH<br>0.5  | MAH<br>0.5                  |
|    | Feed-      | Free radical initiator   | -           | -                  | -                           | _            | -           | -                           |
| 20 | 1          | Paint adhesion improver  | NBR-A<br>5  | ABS-B<br>5         | -                           | PAN<br>5     | MEG<br>5    | -                           |
|    |            | Impact strength improver | Sg-EPR<br>5 | Sg-EPR<br>5        | Sg-EPR<br>10                | mEPR-A<br>5  | mEPR-B<br>5 | mEPR-B<br>5                 |
| 25 |            | Polystyrene              | -           | -                  | -                           | -            | -           | HIPS<br>5                   |
|    |            | Nylon 6                  | 44          | 44                 | 44                          | 40           | 40          | 40                          |
| 30 |            | Nylon 66                 | _           | -                  | -                           | 5            | 5           | 5                           |
|    |            | Aromatic<br>polyamide    | Grivory     | Troga-<br>mid<br>1 | Troga-<br>mid<br>1          | <b>-</b>     | -           | -                           |
| 35 | Feed-<br>2 | Paint adhesion improver  | -           | -                  | -                           | -            | <b>-</b>    | -                           |
| ,  |            | Impact strength          | SBS         | SBS                | SBS                         | mSg-<br>EPDM | mSg-EPF     | mSg-EPR.                    |
| 40 |            | improver                 | 5           | , 5                | 5                           | 5            | 5           | 5                           |
|    |            | Polystyrene              | -           | -                  | -                           | -            | -           | -                           |
| 45 |            | Paint 1 hr               | 19          | 59                 | 0                           | 0            | 0           | 0                           |
|    |            | adhesion 2 hr            | 48          | 60                 | 0                           | 0            | 0           | 0                           |
|    |            | strength 5 hr            | 72          | 86                 | 40                          | 13           | 7           | 0                           |
| 50 |            | 12 hr                    | 89          | 91                 | 61                          | 47           | 29          | 0                           |
|    |            | 24 hr                    | 95          | 99                 | 80                          | 81           | 74          | 11                          |

| PPE-A  |     |        |                       |      |      |            |      |      |
|--|-----|--------|-----------------------|------|------|------------|------|------|
| PPE-A  | •   |        |                       |      |      |            |      |      |
| Compatibilizer   Pumaric acid acid acid acid acid acid acid ac   | 5 • | -      | PPE-A                 | · _  |      | -          | -    | -    |
| Feed-1   Free radical  |     |        | PPE-B                 | 40   | 40   | 40         | 40   | 40   |
| initiator Paint adhesion improver  | 10  |        | Compatibilizer        | acid | acid | acid       | acid | acid |
| Impact strength improver 10 10 10 10 10 10 10 10 10 10 10 10 10  |     | Feed-1 |                       | -    | -    | -          | -    | -    |
| improver 10 10 10 10 10 10 10  Polystyrene   |     |        |                       |      |      |            |      |      |
| Nylon 6  | 20  |        |                       |      |      |            | 1 1  |      |
| Nylon 66   |     |        | Polystyrene           | -    | -    | -          | -    | -    |
| Aromatic polyamide  Feed-2 Paint adhesion  | 25  |        | Nylon 6               | 45   | 45   | 45         | 45   | 45   |
| Peed-2 Paint adhesion  |     |        | Nylon 66              | -    | -    | -          | -    | _    |
| improver  Impact strength  | 30  |        |                       | -    | -    | -          | _    | -    |
| Polystyrene  |     | Feed-2 |                       | -    | -    | -          | -    | -    |
| Paint 1 hr 23 21 11 5 0  adhesion 2 hr 52 50 38 23 0  strength 5 hr 71 72 58 48 25  12 hr 89 88 75 61 41  24 hr 97 96 92 85 59 | 35  |        |                       | -    | -    | -          | -    | -    |
| Paint 1 hr 23 21 11 5 0 adhesion 2 hr 52 50 38 23 0 strength 5 hr 71 72 58 48 25 12 hr 89 88 75 61 41 24 hr 97 96 92 85 59     |     |        | Polystyrene           |      | -    | <b>-</b> . | -    | _    |
| strength 5 hr 71 72 58 48 25  12 hr 89 88 75 61 41  24 hr 97 96 92 85 59   | 40  |        | Paint 1 hr            | 23   | 21   | 11         | 5    | o    |
| 12 hr 89 88 75 61 41<br>24 hr 97 96 92 85 59   |     |        | adhesion 2 hr         | 52   | 50   | 38         | 23   | 0    |
| 12 hr 89 88 75 61 41<br>24 hr 97 96 92 85 59   | 45  |        | strength 5 hr         | 71   | 72   | 58         | 48   | 25   |
| 50   |     |        | 12 hr                 | 89   | 88   | 75         | 61   | 41   |
|  |     |        | 24 hr                 | 97   | 96   | 92         | 85   | 59   |
| discoloration \( \sum_{\text{\text{\text{\text{\text{discoloration}}}}} \)   | 50  |        | Heat<br>discoloration | X    |      | 0          | 0    | 0    |

|            |                         | EX<br>23           | EX<br>24           | Compar-<br>ative<br>EX<br>8 | EX<br>25       | Compar-<br>ative<br>EX<br>9 |
|------------|-------------------------|--------------------|--------------------|-----------------------------|----------------|-----------------------------|
|            | PPE-A                   | 44                 | 44                 | 50                          | 46             | 50                          |
|            | PPE-B                   | -                  | -                  | -                           | -              | -                           |
|            | Compatibilizer          | MAH<br>0.5         | MAH<br>0.5         | MAH<br>0.5                  | MAH<br>0.5     | MAH<br>0.5                  |
| Feed-      | Free radical initiator  | 0.01               | 0.01               | 0.01                        | , <del>-</del> | -                           |
|            | Paint adhesion improver | SAN-2<br>6         | SAN-3<br>6         | -                           | SAN-2<br>6     | -                           |
|            | Impact strength         | m-EPDM-<br>A<br>10 | m-EPDM-<br>A<br>10 | m-EPDM-<br>A<br>10          | _              | -                           |
|            | Inroganic<br>filler     | -                  | -                  | <b>-</b>                    | talc<br>5      | talc<br>5                   |
|            | Nylon 6                 | 40                 | 40                 | 40                          | 40             | 40                          |
|            | Nylon 66                | -                  | -                  | -                           | -              | _                           |
|            | Aromatic polyamide      | -                  | _                  | -                           | -              | -                           |
| Feed-<br>2 | Paint adhesion improver | -                  | -                  | _                           | -              | -                           |
|            | Impact strength         | _                  | -                  | _                           | -              | -                           |
|            | Polystyrene             | -                  | -                  | -                           | -              | -                           |
|            | Paint 1 hr              | 62                 | 65                 | 0                           | 60             | 0                           |
|            | adhesion 2 hr           | 72                 | 99                 | 0                           | 98             | 0                           |
| )          | strength 5 hr           | 99                 | 100                | 0                           | 100            | 10                          |
|            | 12 hr                   | 100                | 100                | 10                          | 100            | 10                          |
| 5          | 24 hr                   | 100                | 100                | 20                          | 100            | 20                          |

#### Claims

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- 1. A thermoplastic composition comprising,
  - A) from 5 to 95 weight % of at least one polyphenylene ether,
  - B) from 95 to 5 weight % of at least one polyamide,
  - C) paint adhesion improving amount of at least one paint adhesion improver selected from
    - (a) homopolymers or copolymers of unsaturated nitrile compounds and
- (b) homopolymers or copolymers of alkyl(meth) acrylate, excluding an ethylene alkylacrylate copolymer having 50 weight % or higher of ethylene,
- D) from 0.01 to 30 parts by weight of at least one compatibilizer, based on 100 parts by weight of the total of the polyphenylene ether and polyamide.
- 2. A composition according to Claim 1 wherein the paint adhesion improver is incorporated in an amount from 0.5 to 30 parts by weight based on the total of the polyphenylene ether and polyamide.
- 3. A composition according to Claim 1 or Claim 2 wherein the unsaturated nitrile compounds are acrylonitrile and/or methacrylonitrile.
- 4. A composition according to any one of Claims 1 to 3 wherein polyphenylene ether is poly(2,6-dimethyl-1,4-phenylene) ether or a copolymer of 2,6-dimethylphenol and 2,3,6-trimethylphenol or a combination thereof.
  - 5. A composition according to any one of Claims 1 to 4 wherein polyamide is polyamide 6, polyamide 66, a blend of polyamide 6 and polyamide 66, an amorphous polyamide, a blend of polyamide 6 and an amorphous polyamide or a blend of polyamide 66 and an amorphous polyamide.
  - 6. A composition according to any one of Claims 1 to 5 wherein the paint adhesion improver is styrene acrylonitrile copolymer (SAN), styrene and acrylonitrile grafted polybutadiene (ABS), an acrylonitrile and at least one alkyl(meth)acrylate copolymer, polymethylmethacrylate, a copolymer or terpolymer among alkyl-(meth)acrylates, an acrylic rubber having a crosslinkable comonomer therein or a combination of an acrylonitrile polymer and an alkyl(meth)acrylate polymer.
    - 7. A substrate fabricated from a thermoplastic composition according to any one of Claims 1 to 6.

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- 54) Thermoplastic resin composition.
- (57) A novel thermoplastic composition having improved paint adhesion strength comprising,
  - A) from about 5 to 95 weight % of at least one polyphenylene ether.
  - B) from about 95 to 5 weight % of at least one polyamide,
  - C) paint adhesion improving amount of at least one paint adhesion improver selected from the class consisting of
    - a) homopolymers or copolymers of unsaturated nitrile compounds and
    - b) homopolymers or copolymers of alkyl (meth) acrylate, excluding an ethylene alkylacrylate copolymer having about 50 weight % or higher of ethylene,
  - D) from about 0.01 to 30 parts by weight of at least one compatibilizer, based on 100 by weight of the total of the polyphenylene ether and

polyamide.



# EUROPEAN SEARCH REPORT

Application Number

EP 90 30 4238

| D          | OCUMENTS CONS   |   |   |                                 |  |
|------------|---|---|---|---------------------------------|--|
| Category   |   | th indication, where appropriate,<br>evant passages |   | levant<br>claim                 | CLASSIFICATION OF THE<br>APPLICATION (Int. CI.5) |
| x          | EP-A-0 236 593 (GENER/<br>* claims 1-18; examples *   | AL ELECTRIC COMPANY)                                | 1-7   |                                 | C 08 L 71/12<br>C 08 L 77/00<br>C 08 K 5/09      |
| Х          | EP-A-0 244 090 (JAPAN \$<br>* claims 1,22-24; examples  |   | 1-7   |                                 |  |
| D,X        |   | AL ELECTRIC COMPANY) 6, line 7; claims 1-65; exampl | es 1-7  |                                 |  |
| Α          | EP-A-0 184 151 (BASF AC<br>* claims 1-4; examples *   | <br>  | 1-7   |                                 |  |
|            |   |   | <b>3</b>                                      |                                 |  |
|            |   |   |   |                                 | TECHNICAL FIELDS<br>SEARCHED (Int. Cl.5)         |
|            |   |   |   |                                 | C 08 L<br>C 08 K                                 |
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|            | The present search report has   | been drawn up for all claims                        | $\blacksquare$                                |                                 |  |
|            |   |   | <u> </u>                                      |                                 | Evaminor   |
|            | Place of search<br>Berlin   | Date of completion of sear<br>15 November 91        | UII   |                                 | Examiner BOEKER R.B.                             |
| Y:  <br>A: | CATEGORY OF CITED DOC<br>particularly relevant if taken alone<br>particularly relevant if combined wit<br>document of the same catagory<br>technological background<br>non-written disclosure | h another D<br>L                                    | the filing da<br>: document c<br>: document c | te<br>Ited in the<br>Ited for o |  |
| P: i       | non-written disclosure<br>Intermediate document<br>theory or principle underlying the in  |   | document                                      | e same                          | parent laming, corresponding                     |